THE EFFECT OF PRESSURE ON THE ANTIFERROMAGNETIC AND SUPERCONDUCTING STATES OF HEAVY-FERMIION UPd$_2$Al$_3$

K. Bakker, A. de Visser, L.T. Tai, A.A. Menovsky and J.J.M. Franse

Van der Waals — Zeeman Laboratorium, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

(Received 13 January 1993 by P. Burlet)

The effect of a hydrostatic pressure ($p \leq 3.5$ bar) on the antiferromagnetic ($T_N = 14.2$ K) and superconducting transition ($T_c = 1.9$ K) temperature of single-crystalline UPd$_2$Al$_3$ has been determined by means of resistivity measurements. The pressure effects are extremely small when compared to other heavy-fermion systems: $dT_N/dp = 0 \pm 3$ mK kbar$^{-1}$ and $dT_c/dp = -2.0 \pm 0.7$ mK kbar$^{-1}$. The effect of a hydrostatic pressure ($p \leq 5$ kbar) on the antiferromagnetic phase boundary has been studied by means of high-field magnetization measurements ($B \leq 28$ T). It is found that the antiferromagnetic phase boundary, which is located at $B_c = 18$ T for a field in the hexagonal plane at $T = 5$ K, shifts to higher field values with pressure at a rate of $0.13 \pm 0.01$ T kbar$^{-1}$.

1. INTRODUCTION

THE OCCURRENCE of superconductivity in a select class of strongly correlated electron systems (CeCu$_2$Si$_2$, UBe$_{13}$, UPt$_3$ and URu$_2$Si$_2$) receives much attention because of the non-standard BCS nature of the superconducting condensate (for a review see e.g. [1–3]). In particular, the presence of pronounced antiferromagnetic spin-fluctuation phenomena, which co-build the heavy quasiparticles, has led to theoretical predictions that an unconventional Cooper state ($L \neq 0$), mediated by an antiferromagnetic electron–electron coupling mechanism, is realized in these systems. Besides, in most of the heavy-fermion superconductors a small-moment antiferromagnetically ordered state coexists with the superconducting state (except in the case of UBe$_{13}$), which raises the question of the interplay of antiferromagnetism and superconductivity. However, this issue is not easily addressed experimentally, not in the least because of the peculiar nature of the antiferromagnetic moment. The relevant experiments performed so far have primarily been successful in correlating the pressure dependences of the superconducting ($T_c$) and antiferromagnetic ($T_N$) transition temperatures.

Recently, the select class of antiferromagnetic heavy-electron superconductors was extended with UNi$_2$Al$_3$ ($T_N = 4.6$ K, $T_c = 1$ K) [4] and UPd$_2$Al$_3$ ($T_N = 14$ K, $T_c = 2$ K) [5], offering new possibilities to investigate the interplay of antiferromagnetism and superconductivity. Both compounds crystallize in the hexagonal PrNi$_2$Al$_3$ structure. Hitherto, single-crystalline samples have only been prepared for the Pd 1:2:3 compound and therefore UPd$_2$Al$_3$ has been characterized rather well experimentally [5–8]. The superconducting and antiferromagnetic phase diagrams show a number of interesting features [8]. As far as the superconductivity is concerned much attention is directed towards the possibility that a Fulde–Ferrell state is realized [9]. As to the antiferromagnetic properties, the fairly large ordered moment of $0.85 \pm 0.03$ $\mu_B$ on the U-atom, detected by neutron diffraction [10], is conspicuous as it contrasts with the small ordered moments that have been reported for the other U-based heavy-fermion superconductors [3]. The magnetic properties are strongly anisotropic with the hexagonal plane as the easy plane for magnetization [6–8]. For $B = 0$, the ordering consists of ferromagnetic sheets parallel to the basal plane which are coupled antiferromagnetically along the hexagonal axis ($c$-axis), i.e. a doubling of the nuclear unit cell with an ordering vector $k = [0, 0, 0.5]$. The antiferromagnetic phase diagram in the $B$–$T$ plane consists of three phases with critical fields of 0.6 T, 4.2 T and 18 T (for $T \to 0$ and fields directed in the basal plane) [7, 8]. The magnetic structures of the field-induced phases are not yet resolved. Recently,
\(\mu SR\) experiments at temperatures as low as 50 mK indicate that antiferromagnetism and superconductivity coexist [11].

In this paper we report on the variation of \(T_c\) and \(T_N\) with hydrostatic pressure as determined by electrical resistivity measurements. Furthermore, we report on high-pressure high-field magnetization measurements which enabled us to study the pressure dependence of the antiferromagnetic phase boundary, which is located at \(B_c = 18\) T at ambient pressure (for \(T \rightarrow 0\)).

2. EXPERIMENTAL

A single-crystalline batch of UPd\(_2\)Al\(_3\) was prepared in a titanium gettered argon atmosphere using a modified tri-arc Czochralski technique. After an annealing procedure (7 days at 900°C) various samples were cut from the single-crystalline batch by means of spark erosion. For the high-pressure resistivity and high-pressure magnetization experiments a bar-shaped specimen was prepared (with dimensions \(1 \times 1 \times 3.5\) mm\(^3\)), with the long direction along the \(a\)-axis. Resistivity experiments performed for a current along the \(a\)-axis yield a residual resistance value \(\rho_0 = 6.2\) \(\mu\)\(\Omega\) cm. The residual resistance ratio, \(R(300\) K)/\(R(0\) K), equals 28. The onset for superconductivity occurs at \(T_c^{onset} = 1.88\) K. The width of the superconducting transition is rather broad (0.2 K), which is ascribed to small variations in the aluminium concentration. Recently, single-crystalline material prepared with excess aluminium (nominal composition UPd\(_2\)Al\(_{1.03}\)) [12], was found to exhibit a much sharper resistive superconducting transition. Possibly, excess starting material compensates for the evaporation of aluminium during the crystal growth procedure.

For the resistivity experiments under pressure the sample was mounted on a support with current and potential leads attached to it by means of soldering. The support and sample were placed in a pressure vessel (cylindrical shape, outer diameter 25 mm, inner diameter 6 mm), constructed of a copper–beryllium alloy. The high-pressure low-temperature electrical feedthrough was realized by embedding the wires in epoxy and gluing them on a conically machined plug that served as pressure sealing. Helium was used as pressure transmitting medium. The pressure cell was mounted in a cylindrical pressure vessel with strongly reduced dimensions (outer diameter 5 mm, inner diameter 1.7 mm). The vessel was machined of a copper–beryllium alloy. Again helium was used as the pressure transmitting medium. The pressure cell with sample was placed in a compensated pick-up coil system. The magnetization was measured by integrating the induced voltage. In the high-field installation pulsed magnetic fields can be generated up to 40 T, with a total pulse time of about 1 s. The energy is taken directly from the mains. Different pulse shapes can be chosen as the field can be regulated by a thyristor rectifier that is coupled to the mains. An experimental difficulty when using metallic pressure vessels in pulsed magnetic fields at low temperatures is the generation of heat due to the dissipation of eddy currents. As the pressure-transmitting solid helium is a good thermal conductor, the temperature of the sample might change. In order to keep the temperature raised of the sample within acceptable limits, a low field sweep rate of 30 T s\(^{-1}\) was chosen. Furthermore, the pressure vessel and pick-up coil were immersed directly in liquid helium (at atmospheric pressure). The effective temperature of the sample during the magnetic field pulse amounts to 5.0 ± 0.3 K. The data at various pressures have been taken for increasing fields, using identical field profiles. The as-measured signal was corrected for the remaining unbalance of the pick-up coil system and the signal from the empty pressure vessel. In the high-field range the magnetization of the sample exceeds that of the pressure cell by a factor 5.

3. RESULTS AND ANALYSIS

The electrical resistivity of UPd\(_2\)Al\(_3\) for a current along the \(a\)-axis at ambient pressure for \(T < 300\) K is shown in Fig. 1. The increase in \(\rho(T)\) with decreasing temperature is ascribed to a mixture of Kondo and crystalline electric field effects, yielding a broad maximum centred at 80 K. At low temperatures, in the coherent state, a pronounced kink is observed, maintaining a temperature gradient, as to start solidification at the bottom of the cell. The pressure was monitored by strain gauges that were glued on the pressure vessel.

The magnetization experiments under pressure were performed in the High Magnetic Field Facility of the University of Amsterdam [13]. The sample was mounted in a cylindrical pressure vessel with strongly reduced dimensions (outer diameter 5 mm, inner diameter 1.7 mm). The vessel was machined of a copper–beryllium alloy. Again helium was used as the pressure transmitting medium. The pressure cell with sample was placed in a compensated pick-up coil system. The magnetization was measured by integrating the induced voltage. In the high-field installation pulsed magnetic fields can be generated up to 40 T, with a total pulse time of about 1 s. The energy is taken directly from the mains. Different pulse shapes can be chosen as the field can be regulated by a thyristor rectifier that is coupled to the mains. An experimental difficulty when using metallic pressure vessels in pulsed magnetic fields at low temperatures is the generation of heat due to the dissipation of eddy currents. As the pressure-transmitting solid helium is a good thermal conductor, the temperature of the sample might change. In order to keep the temperature raised of the sample within acceptable limits, a low field sweep rate of 30 T s\(^{-1}\) was chosen.

Furthermore, the pressure vessel and pick-up coil were immersed directly in liquid helium (at atmospheric pressure). The effective temperature of the sample during the magnetic field pulse amounts to 5.0 ± 0.3 K. The data at various pressures have been taken for increasing fields, using identical field profiles. The as-measured signal was corrected for the remaining unbalance of the pick-up coil system and the signal from the empty pressure vessel. In the high-field range the magnetization of the sample exceeds that of the pressure cell by a factor 5.
The sudden drop of $\rho(T)$ at $T_N$ for UPd$_2$Al$_3$ contrasts with the increase in resistivity just below $T_N$ as observed for the heavy-fermion antiferromagnets URu$_2$Si$_2$ ($T_N = 17.5$ K) [15] and U(Pt$_{0.95}$Pd$_{0.05}$)$_3$ ($T_N = 5.8$ K) [16]. In the latter compounds the antiferromagnetic transition of the spin-density wave type is accompanied by the formation of a gap over a part of the Fermi surface. In the case of URu$_2$Si$_2$, the normal-state electrical resistivity below $T_N$ could satisfactorily be analyzed using the expression for an antiferromagnet with an energy gap, $\Delta$ (of the order of 80 K), and a $T^2$-term due to the Fermi-liquid behaviour [15]:

$$\rho = \rho_0 + BT[1 + 2T/\Delta] \exp(-\Delta/T) + AT^2.$$  

Figure 4 shows that the $\rho(T)$-data ($I \parallel a$) of UPd$_2$Al$_3$ are described reasonably well using equation (1). The optimum fit parameters are: $\rho_0 = 6.2 \mu\Omega$ cm, $B = 30 \mu\Omega$ cm K$^{-1}$, $\Delta = 39$ K and $A = 0.24 \mu\Omega$ cm K$^{-2}$. Recently, the resistivity of polycrystalline UPd$_2$Al$_3$ was analyzed in a similar way by Dalichaouch et al. [14], yielding $\Delta = 40$ K. Hence, the resistivity data suggest that also in UPd$_2$Al$_3$ a gap is formed at $T_N$. On the other hand, close to $T_N$, small deviations from equation (1) appear, while also the typical increase in $\rho(T)$ below $T_N$ due to the decrease of carriers is absent. A careful analysis of the electronic specific heat, including an
exponential term, might further elucidate this point. A fit of the ρ(T) data under pressure to equation (1) yields identical parameters ρ0, A, B and Δ within the accuracy of the analysis.

Next, we present the results of the high-pressure high-field magnetization experiments. At ambient pressure at T = 4.2 K the magnetization of UPd2Al3 for a field in the hexagonal plane shows a sharp jump at Bc = 18.0 T [7]. As the size of the jump (ΔM = 0.92 μB/U-atom) is almost equal to the size of the ordered moment (0.85 μB/U-atom), the transition at Bc most likely reflects the antiferromagnetic phase boundary. Under pressure, Bc shifts to higher fields as is shown in Figs. 5 and 6. In the pressure range 1 kbar < p < 4.9 kbar, Bc increases linearly with pressure at a rate dBc/dp = 0.13 ± 0.01 T kbar⁻¹. At smaller pressures the variation of Bc is nonlinear. The variation of the magnetization with pressure, M(p), above and below Bc, cannot be determined from the present experiments, as it falls within the experimental uncertainty (0.1 μB/U-atom). The overall effect on the M(B) curve is very small for the applied pressures. Also, the pressure variation of the size of the jump at Bc is negligibly small: dΔM/dp = 0 ± 0.4 × 10⁻² (μB/U-atom) kbar⁻¹.

4. DISCUSSION

In general, the low-temperature properties of heavy-fermion systems are extremely sensitive to pressure, because of the strong hybridization of the f-orbitals of Ce or U with the p or d-orbitals of the ligand atoms. Consequently, large Grüneisen parameters are found for the volume variation of the characteristic temperature, T∗, below which the heavy quasiparticle bands are formed: Γ = -∂ln T*/∂ln V ≈ 100 [17]. As the heavy quasiparticles participate in the formation of the superconducting and antiferromagnetic states, also Tc and TN depend strongly on pressure, e.g. for UPt3: Γsc = -∂ln Tc/∂ln V = -52 [18], for U(Pt0.95Pd0.05)3: Γaf = -∂ln TN/∂ln V = -110 [19] and for URu2Si2: Γsc = -62 and Γaf = 16 [20] (note that in the case of URu2Si2 the Grüneisen parameters are calculated with an estimated compressibility κ = 0.73 Mbar⁻¹). These large values clearly contrast with the small values observed for UPd2Al3: Γsc = -1 ± 0.4 and Γaf = 0 ± 0.2 (here we used an estimated compressibility κ = 1 Mbar⁻¹). As the pressure dependence of Tc is concerned, the small value of Γsc is in agreement with the near absence of a discontinuity at Tc in the coefficients of thermal expansion, measured for a single-crystalline sample along (α∥) and perpendicular to (α⊥) the hexagonal axis [6] (conform the Ehrenfest relation). Note that Γaf has not been determined reliably yet via thermal expansion, as data have only been obtained for a polycrystalline sample with preferentially oriented crystallites [6].

Apparently, superconductivity and antiferromagnetism in UPd2Al3 are correlated in the sense that both are rather insensitive to pressure. It is interesting to note that the very weak pressure dependence of Tc (and probably also of TN) does not result from a near cancellation of the uniaxial pressure effects, as α∥ and α⊥ show no large discontinuities at Tc. Hence, both uniaxial and hydrostatic pressure effects are small. This strongly suggests that the hybridization effects in UPd2Al3 are less important than in other heavy-fermion compounds and, consequently, a more localized
character of the f-electrons is inferred. This claim is supported by the relatively large size of the ordered moment.

Although $T_N$ is almost pressure independent, the low-temperature antiferromagnetic phase boundary shifts upwards at a rate $dB_c/dp = 0.13 \text{TKbar}^{-1}$, indicating a stronger antiferromagnetic coupling under pressure. The actual magnetization process, with the hexagonal plane as easy plane for magnetization, is far from being unravelled. The zero-pressure high-field magnetization [7] and magnetoresistance [21] experiments yield one sharp transition at $B_c = 18\text{T}$, which reflects the antiferromagnetic phase boundary, and the absence of a marked basal-plane anisotropy. However, in a further detailed investigation a small basal-plane anisotropy was detected [22]. This possibly suggests that the transition at $B_c$ is of the spin-flip type. Furthermore, recent magnetization, magnetostriction and magnetoresistance experiments have revealed the presence of two additional phase lines (with critical fields of 0.6T and 4.2T for $T \to 0$) indicating that the paramagnetic state is reached via two field-induced magnetic phases [8]. Neutron-diffraction experiments in a magnetic field would be most welcome to elucidate the magnetic structures of the various phases.

In conclusion, we have studied the pressure dependence of the superconducting ($T_s$) and antiferromagnetic ($T_N$ and $B_c$) phase boundary of UPd$_2$Al$_3$ via resistivity and magnetization experiments. The deduced pressure variations are much smaller than observed for other heavy-fermion compounds, suggesting a dominant local character of the f-electrons.

Acknowledgements — The research of one of us (AdV) was made possible by a fellowship of the Royal Netherlands Academy of Arts and Sciences. S.A.M. Mentink is gratefully acknowledged for a first characterization of the sample. The authors thank R. Scheltema for skilful assistance during the experiments.

REFERENCES
